ISSN 0704-3716

Canadian Translation of Fisheries and Aquatic Sciences

No. 5541

The characteristics and distribution of polycyclic aromatic hydrocarbons in sea sediments in the Kitakyushu area

Y. Hanada, S. Ishikawa, S. Sueta, and K. Kido

Original title: Kitakyushu-shi Shuhen Kaiiki Teishitsuchu no Takanhokozoku Tankasuiso no Nodo Bunpu to Tokucho

In: Eisei Kagaku/[Health Science] 36(1): 8-14, 1990

Original language: Japanese

Available from:
Canada Institute for Scientific and Technical Information
National Research Council
Ottawa, Ontario, Canada K1A 0S2

1991

15 typescript pages

Secretary of State / Secrétariat d'État

MULTILINGUAL SERVICES DIVISION – DIVISION DES SERVICES MULTILINGUES

TRANSLATION BUREAU / BUREAU DES TRADUCTIONS

92-00830/

LIBRARY IDENTIFICATION – FICHE SIGNALÉTIQUE

Translated from - Traduction de	Into - En
Japanese	English

Author - Auteur

Yoshifumi HANADA, Seiichi ISHIKAWA, Shintaro SUETA and Kozo KIDO

Title in English or French – Titre anglais ou français

The Characteristics and Distribution of Polycyclic Aromatic Hydrocarbons in Sea Sediments in the Kitakyushu Area

Title in foreign language (Transliterate foreign characters)
Titre en langue étrangère (Transcrire en caractères romains)

Kitakyushu-shi Shuhen Kaiiki Teishutsuchu no Takanhokozoku Tankasuiso no Nodobunpu to Tokucho

Reference in foreign language (Name of book or publication) in full, transliterate foreign characters.
Référence en langue étrangère (Nom du livre ou publication), au complet, transcrire en caractères romains.

Eisei Kagaku

Reference in English or French - Référence en anglais ou français

Health Science

Publisher - Éditeur	DATE OF PUBLICATION / DATE DE PUBLICATION			Page Numbers in original / Numéros des pages dans l'original
	Year / Année	Volume	Issue No. / Numéro	8 - 14
Place of Publication / Lieu de publication	1990	36	1	Number of typed pages / Nombre de pages dactylographiées: 14

Requesting Department / Ministère-Client: DFO

Branch or Division / Direction ou Division: Scientific Pubs / Comm. Dir.

Person requesting / Demandé par: V. Zitko – Biological Station, St. Andrews, New Brunswick, EOG 2X0

Your Number / Votre dossier nº:

Date of Request / Date de la demande: July 23, 1991

Translation Bureau No. / Notre dossier nº: 3850549

Translator (Initials) / Traducteur (initiales): PS

AUG / AOUT 14 1991

UNEDITED TRANSLATION
For information only
TRADUCTION NON REVISEE
Information seulement

Canada

SEC 9-111 (84-10)

UNEDITED TRANSLATION
For information only
TRADUCTION NON REVISEE
Information seulement

Secretary of State—Secrétariat d'État

MULTILINGUAL TRANSLATION DIRECTORATE
DIRECTION DE LA TRADUCTION MULTILINGUE

TRANSLATION BUREAU
BUREAU DE LA TRADUCTION

Client's No. – N° du client	Department – Ministère	Division/Branch – Division/Direction	City – Ville
	DFO	Scientific Publications/Communications Dir.	Ottawa
Bureau No. – N° du Bureau	Language – Langue	Translator – Traducteur	AUG AOUT 14 1991
3850549	Japanese	PS	

The Characteristics and Distribution of Polycyclic Aromatic Hydrocarbons in Sea Sediments in the Kitakyushu area

(Kitakyushu-shi Shuhen Kaiiki Teishitsuchu no Takanhokozoku Tankasuiso no Nodo Bunpu to Tokucho)

by Yoshifumi HANADA, Seiichi ISHIKAWA, Shintaro SUETA and Kozo KIDO

Kitakyushu Municipal Institute of Environment Health Sciences, Shin-ike 1-2-1, Tobata-ku, Kitakyushu 804, Japan

(received June 8, 1989)

in **Eisei Kagaku** (Health Science), Vol. 36, (1990), pp. 8 - 14

Foreword

Most polycyclic aromatic hydrocarbons (PAH) are produced at the burning stage of fossil fuels. As they are released in the environment, they spread widely throughout the environment which includes atmosphere and hydrosphere.[1] Moreover, they are contained in the fossil fuels themselves, and have been detected in various types of wastewater such as factory effluents and city sewage.[2] Therefore, large amounts have been detected in the environment of urban and industrial regions where there is much human activity.[3] The discharged PAHs run into the rivers and sea, and move to the sediments where they accumulate. It seems therefore that the analysis of the PAHs in sediments should yield results that could be used as the pollution indeces of the particular pelagic areas. Kitakyushu city is an industrial city which developed from the late 19th century, mainly for activities related to heavy

chemical industry using coal and petroleum, and it seems that the pollution caused by the combustion of fossil fuels is worsening. This is why we performed quantitative analyses of the PAHs in the sea sediments of three different pelagic areas, that is, the Dokai Bay which is a semi-enclosed area,

p. 9

the Kannon strait where the tidal current is violent, and the Sea of Suo where there is a strong sedimentary rocks action. The purpose was to know the actual state of pollution brought about by fossil fuels. Moreover, on the basis of the results obtained through a multivariate analyses of the PAH compositions in the sediments, we examined the particularities of the regions, registering the similarities and characteristics of the particular sites. In those sediments where the presence of PAH had been confirmed, the following 14 species were detected in relatively large quantities, that is, phenanthrene (Phe), anthracene (An), fluoranthene (Fl), pyrene (Py), benzo [a] fluorene (B[a]F), benzo [*b*] fluorene (B[*b*]F), benzo [a] anthracene (B[a]A), chrysene (Ch) + triphenylene (Tr), benzo [*b*] fluoranthene (B[*b*]Fl), benzo [e] pyrene (B[e]P), benzo [a] pyrene (B[a]P), perylene (Per), indeno [1, 2, 3-cd] pyrene (Ind), benzo [*ghi*] perylene (B[*ghi*]p).

Survey and testing method

1. Survey sites and samples

The sites of survey are shown in Fig. 1.

The survey was centered on the Sea of Suo region where in spite of the environment having become cleaner over the recent year, the environmental standards (rank A : not more than 2 ppm COD) have not been met yet. We also surveyed the sediments of the coastal region so as to examine the effect of the land on the Sea of Suo. We collected samples from March to October 1988. For the coastal region, we collected surface samples (10 cm) with a transplanting iron. For the pelagic areas, we used an Ekman-Berge mud collector and analyzed the samples after storage in a dark and cool place.

2. The reagents

For Phe, An, Fl, Py, B[a]P and Per we used reagents of the Kanto Chemical Co, for B[a]F, B[*b*]Fl and B[e]P we used reagents of the Wako Jun'yaku

Industries, for B[*b*]F and B[a]A we used reagents of the Tokyo Kasei Industries, and for Ch, B[*ghi*]P we used reagents of the Aldrich Co. As for the other reagents, we distilled commercial products made for residual pesticide, or top-grade preparations.

3. Method of analyzing the PAH

Although numerous methods of analyzing PAH have been reported,[1-3] there exists no established method. We performed extraction with acetone[4] as it has a great power for extracting organic matters in sediments. After performing clean-up with the Sep-pak silica cartridge reported by Kawada et al [5], we used a GC-MS capillary column as it has outstanding selectivity and sensitivity. By doing so, we obtained excellent PAH analytical results. We show hereunder the method of proceeding and the results of the recovery tests.

3 - 1 Proceeding method

We shake-extracted 5 - 50 g of sediment sample (wet weight) twice, using 100 mL of acetone per extraction, transferred the extracted acetone solution to a separatory funnel, added 100 mL of dichloromethane, following which we washed twice, using 200 mL of pure water per washing. We concentrated the dichloromethane layer with a Kuderna-Danish concentrator but added 10 mL of isooctane during concentration, and concentrated finally so as to have a 2 mL isooctane solution. We performed the clean up of the concentrate with a Sep-pak silica cartridge (from the Waters Co.). If there is much deposit in the concentrate, and if the deposit becomes attached to the container, the concentrate is passed through the Sep-pak silica cartridge, after which the deposits remaining in the container are dissolved with 50 µL of dichloromethane. Following this, we added 1 mL of isooctane and allowed it to adsorb on the Sep-pak silica cartridge through which the concentrate had been passed. We performed the elution from the Sep-pak silica cartridge with 8 mL

of a isooctane : benzene solution (1 : 1). We concentrated this eluate to 2 mL with nitrogen gas, and after adding an internal standard (*p*-terphenyl - d 14), analyzed by GC-MS. The GC-MS equipment consisted of a 5890 A model GC-MS Hewlett-Packard unit connected to a DX 303 model from Nippon Denshi. The measurement conditions are given in Table I.

3.2 Recovery test

We added 50 μg each of PAH to 50 g of sediment from 1 site (S 2) of the Sea of Suo region, and performed the measurements according to the operasting method. For control, we analyzed similarly sediment to which no PAH had been added, and measured the recovery rate. The results are shown in Table II. Fig. 2 shows the SIM chromatogram of mud to which PAH had been added.

As a result, all PAH indicated a recovery rate of at least 80 %, and we were able to eliminate the interfering substances by passage through the Sep-pak silica cartridge.

Results and discussion

1. The distribution of PAH concentration

Table III shows the PAH quantitative results obtained at 24 sites around the city of Kitakyushu and the coastal area of the Sea of Suo. The 14 targetted species of PAH were detected in the sediments at all measurement sites.

The calculations of the totals of the 14 species of PAH quantified as total PAH (T.PAH) revealed higher tendencies in the pelagic regions as the pelagic areas indicated 0.52-36.6 μg/g, and the coastal area of the Sea of Suo 0.083 - 0.796 μg/g. The D2 site in the pelagic area of the Dokai Bay showed the highest value with 36.6 μg/g, followed by site D1 with 35.8 μg/g. Site D3 and site K1 in the Kannon strait showed also higher values with 5.57 and 6.60 μg/g respectively. Sites D1 and D2 near the mouth of the Dokai Bay had higher

p. 12

concentrations than site D3 at the rear of the bay, and there seems to be a local source of pollution near the mouth of the bay. Searl et al. reported that ppb-levels of B[a]P and other PAHs had been detected in water discharged from coke manufacturing plants.[6] We analyzed the water discharged from one coke manufacturing plant located near the Dokai Bay and detected all 14 target PAH species. The T.PAH concentration was 37.4 μg/L. When calculating the amount of negative load from this discharge mouth on the basis of the wastewater volumes, we discovered that the T.PAH was 2.6 Kg, and that a rather large volume of PAH was being discharged. The region bordering on the proximity of the Dokai Bay and on the western part of the Kannon strait is the industrial belt of the city of Kitakyushu, and has numerous coke ovens as well

as places of storage. One is therefore expecting at the S1, D2, D3 and K1 sites not only exhaust gas from the fossil fuels used at the plants but a strong effect of the waters discharged from plants related to fossil fuels as well.

The maximum T.PAH concentration in the sediments of the Sea of Suo region is 1.80 µg/g. The level was lower than in the Dokai Bay or in the Kannon strait. In the northern part (S1, S2, S5, S8 and S9) of the Sea of Suo region, the concentration was 0.91 - 1.80 µg/g. The PAH accumulation was about double that observed in the southern part (S3, S4, S6, S7 and S 10) which indicated 0.52 - 0.88 µg/g.

The T.PAH concentration in the coastal part of the Sea of Suo (E1-E10) was 0.083-0.796 µg/g and lower than in the pelagic region of the Sea of Suo. However when considering the ignition loss (IL), we note in the coastal region of the Sea of Suo large differences in the volumes of organic matters contained in the sediments. PAH adsorption on the sediments depends on the amount of organic matters in the sediments [7]. Therefore, the T.PAH concentration per weight of sediment does not reflect precisely the state of PAH accumulation at each site. So, if we calculate and compare the volumes of T.PAH per IL, we note 0.7-15.6 µg/g for the coastal areas, 9.0-32.1 for the pelagic areas and a tendency to lower values in the coastal regions.

It is therefore estimated that in the pelagic region of the Sea of Suo there is little PAH contamination effect from the land.

With regard to the concentration of B[*a*]P and B[*e*]P, which is its isomer and which is confirmed to be carcinogenic, we made comparisons with the Tokyo Bay.[8] In the Tokyo Bay, the total was 0.038-0.173 µg/g but in the Dokai Bay (D1-D3), it was 1.0-7.1 µg/g. It was 1.1 µg/g in the Kannon strait (K1), 0.087-0.31 µg/g in the Sea of Suo (S1-S10). For all pelagic regions, the values tended to be higher than in the Tokyo Bay.

2. Study of the state of contamination, based on PAH pattern analysis

From the T.PAH values at each site we obtained a general view of the state of sediment contamination by the PAHs. We examined in detail the state of contamination of the neighboring pelagic regions by further analyzing the PAH compositions of the three pelagic regions. All 14 PAH species were detected at every survey point in the Dokai Bay (D1-D3), Kannon strait (K1) and Sea of Suo (S1-S10). B[*b*]F1 had the highest value, and no great

differences in PAH composition were noted between the sites. Using the relative proportions of the 14 PAH species as characteristic values, we performed cluster analyses[9] as well as analyses of the principal components [10].

In the cluster analysis, we calculated the distance indices on the basis of the standard Euclidian distances. Thereafter, we performed systematic classification according to the group average method in which the distances between the clusters are maintained even when converting to clusters in the stratum method. Fig. 3 shows the analytical results in a dendrogram fashion.

If on the basis of the results of Fig. 3 one classifies the various sites of the pelagic areas according to 4 clusters, we obtain the following groupings, that is, the Sea of Suo pelagic region excepting S8 (group A), the sites D1 and D2 (group B), and the sites D3 and K1 (group C). The T.PAH concentrations of the various groups were : 0.52-1.80 μg/g for group A, 35.8-36.6 μg/g for group B, and 5.57-6.00 μg/g for group C. We noted the same concentration levels of contamination for each group. The results of the cluster analysis show that the presently surveyed relative proportions of PAH in the pelagic sediments reveal resembling patterns in those areas where the concentration levels of PAH contamination are the same. These findings suggest that the types of contamination sources or contamination routes vary according to the sites, and that valuable information could be obtained for future elucidation of pollution sources and for a knowledge of the regional

p. 13

characteristics. As an example, we calculated the standard Euclidian distances[9] and compared the PAH pattern similarities of the groups divided through the cluster analysis and of the water discharged by the coke manufacturing plants. With regard to the water discharged by the coke-manufacturing plants, the value of group B (D1, D2) was smallest at 17.3. For the groups A and C, the values were 22.9 and 59.3 respectively. This indicates that the PAH patterns of the sites D1, D2 whose local pollution was observed in the former section are more like those of the waters discharged by the coke-manufacturing plants than those of the other sites. It is estimated that the waters discharged by the coke manufacturing plants affect strongly the pollution at the former sites.

Furthermore, as in the cluster analysis one obtains only for site S8 (which is in the same pelagic area) results differing from the other results

for the Sea of Suo pelagic region, we used the analysis of the principal components to observe the points of difference in the patterns. We performed the analysis, using correlation queues, and taking the relative proportion of PAH at the various sites as characteristic values. In this case, the cumulative contribution rates of the first principal component (Z1) and of the second principal component (Z2) were 0.753. Tables IV and V show the results of the principal component analyses.

If we observe the score for the principal component Z1, we see that D1, D2 have the respective close values of 0.723 and 0.728. So, like in the results obtained regarding the T.PAH concentrations and the cluster analysis, the sources of pollution are estimated to be identical. The scores for the principal component Z1 at the site S8 is - 1.751, and is within the - 0.728 - -1.912 range for the Sea of Suo pelagic region. No characteristic is revealed by Z1. Yet, with the scores for the principal component Z2, the site S8 tended toward the highest negative value of - 3.348. If we consider the amount of factor negative load of Z2, we see that B[*b*]Fl with the molecular weight of 252 represents the positive-negative borderline. This indicates that Z2 is a principal component whose characteristic is to tend toward the negative when the proportion of low molecular weight side increases from Phe with the molecular weight of 178 to Ch + Tr with the molecular weight of 228, and to tend toward the positive when the proportion of high molecular weight increases from B[*b*]Fl with the molecular weight of 252 to B[*ghi*]P with the molecular weight of 276. Consequently, the PAH pattern of the S8 site shows that the proportion of PAH on the low molecular weight side in the Sea of Suo pelagic region is highest. The report by In et al [8] suggests that the relative proportion of PAHs with low molecular weight vs. T.PAH decreases as it moves to the water, to the floating precipitated particles and sediments, and that the relative proportion of PAH with low molecular weight in the sediments decreases as the distance from the river mouth increases in the case of waters and the like discharged by plants and flowing into the pelagic areas via the rivers. If, assuming that the PAH pollution in the sediments of the Sea of Suo pelagic area proceeds mainly as an aqueous system, one compares this view with the observations regarding site 8S, we see that the inflow of PAH into the Sea of Suo pelagic area proceeds in a large proportion by going first through site S8. Site S8 is among the survey sites in the Sea of Suo the site located to the fartest northeast, and is remote from the coast of

Kitakyushu city. The fact that the flow of PAHs into the Sea of Suo pelagic area goes first through site S8 agrees with "this pelagic region is little affected from the coastal area of Kitakyushu city" in the former section.

Conclusion

We measured the PAH in the pelagic regions and in the coastal sediments around the city of Kitakyushu, and obtained the following results.

p. 14

(1) The T.PAH concentration is 5.57-36.6 µg/g in the Dokai Bay, 6.00 µg/g in the Kannon strait, 0.522-1.80 µg/g in the Sea of Suo, and 0.083-0.796 µg/g on the coast of the Sea of Suo. The values are highest in the Dokai Bay in whose vicinity numerous coke-manufacturing plants and storages are located. The 14 species of PAH were detected at all survey points, and except for one site in the coastal region, the concentration of B[*b*]Fl was always the highest.

(2) The concentration of B[*a*]P + B[*e*]P was 0.087-7.1 µg/g in the sediments of the pelagic areas around the city of Kitakyushu, and all pelagic areas tended to have higher values than in the Tokyo Bay (0.038-0.173 µg/g). One could see the typical features of an industrial city developed mainly for coal and petroleum activities.

(3) We recorded high T.PAH concentrations at 2 sites close to the mouth of the Dokai Bay. On the basis of cluster analyses and of analyses of the principal components, the cause (of pollution) was estimated to be the waters discharged by a coke-manufacturing plant.

(4) The sediments of the Sea of Suo where accumulation tends to be strong showed the lowest T.PAH concentration levels in the pelagic areas surveyed. The fact that the PAH concentrations in the northern pelagic areas were relatively high, and that the concentrations in the coastal sediments on this side of the city were lower than the concentrations in the pelagic sediments suggest that the inflow of PAHs from the coast of the present city has little effect. This was corroborated by the analysis of the main components.

(5) By performing multivaried analyses taking the various PAH relative proportions as characteristic values, we were able to observe in detail the similarities between the pelagic areas and between the survey sites, as well as their characteristics.

(6) Through the analysis of the principal components, we clarified with regard to the PAH in the sediments in the northeast part of the Sea of Suo that there was a large proportion of PAHs with low molecular weight going from Phe with a molecular weight of 178 to Ch + Tr with molecular weights of 228.

Acknowledgment

We thank the personnel of the Water Quality and Noise Section of the Bureau for Pollution Policies of the City of Kitaskyushu who provided us with much help in the course of this study.

Fourteen polycyclic aromatic hydrocarbons (PAH) in sea and seashore sediments in the Kitakyushu district were determined by GC-MS. The similarity and characteristics among the sea areas or sampling sites were defined by the multivariate analysis of PAH composition.

The concentration of total PAH (T · PAH) in the sediments of Dokai Bay, Kanmon Strait, Sea of Suo and the seashore along Sea of Suo were 5.57—36.6, 6.00, 0.522—1.80 and 0.083—0.796 $\mu g/g$ · dry weight, respectively. Fourteen PAH were detected at every sites and the level of benzo[*b*]fluoranthene was the highest of all PAH at every sites except a site of the seashore. The level of benzo[*a*]pyrene + benzo[*e*]pyrene concentration in the 3 sea areas was higher than that in Tokyo Bay. The fact would be characteristic of the industrial city which had developed with coal and petroleum.

In the mouth of Dokai Bay, the value of T · PAH was the highest. This cause was assumed to be an inflow of coke oven effluent by the cluster analysis.

The level of T · PAH concentration in the sea of Suo was the lowest. The ratio of lower molecular weight (178—228) PAH in the northeastern site were relatively higher than that of the other site. The value of T · PAH in the seashore was lower than that in the sea of Suo. By considering these characteristics, it would be assumed that the influence from Kitakyushu side was less than that from the other sides in this sea area. This was confirmed by the principal component analysis.

The interpretation of the results obtained from the multivariate analysis of PAH patterns made it possible to define the characteristics of PAH accumulated in the sea sediments.

Keywords —— polycyclic aromatic hydrocarbon; sea sediment; GC-MS; cluster analysis; principal component analysis

Fig. 1. Location of Sample Sites

TABLE I. Operational Conditions for GC-MS Analysis

GC Condition	
Column	ultra 2 (5% phenylmethylsilicone) 25 m × 0.32 mm i.d.
Column temp.	80°C (1 min) – (30°C/min) – 200°C – (10°C/min) – 300°C
Injector temp.	300°C
Injector mode	splitless
Carrier gas	He
Internal standard	*p*-terphenyl d 14
MS Condition	
Ionization mode	electron ionization (EI)
Ion accelerating voltage	3 kV
Ionization voltage	70 eV
Ionizing current	300 μA
Ion source temp.	280°C
Measurement mode	selected ion monitoring (SIM)

TABLE II. Recoveries of PAH from Sediment

PAH	Mol. wt.	Recovery (%) $n=3$
Phe	178	87±3
An	178	80±4
Fl	202	92±3
Py	202	91±3
B[*a*]F	216	92±3
B[*b*]F	216	92±2
B[*a*]A	228	95±3
Ch+Tr	228	97±2
B[*b*]Fl	252	103±2
B[*e*]P	252	104±4
B[*a*]P	252	97±4
Per	252	96±5
B[*ghi*]P	276	100±7

Each 50 μg of PAH was added to 50 g sediment

TABLE III. Concentrations of PAH in Sediment [μg/g·dry]

Site	Phe	An	Fl	Py	B[a]F	B[b]F	B[a]A	Ch+Tr
D1	1.3	0.94	2.6	3.4	1.3	1.0	2.2	2.8
D2	1.4	0.94	2.8	4.1	1.3	1.1	2.2	2.8
D3	0.33	0.24	0.42	0.54	0.29	0.25	0.28	0.32
K1	0.30	0.21	0.47	0.61	0.27	0.22	0.33	0.40
S1	0.061	0.032	0.15	0.191	0.066	0.041	0.12	0.14
S2	0.035	0.018	0.087	0.11	0.039	0.025	0.072	0.079
S3	0.036	0.017	0.075	0.11	0.033	0.021	0.057	0.059
S4	0.017	0.009	0.041	0.069	0.020	0.013	0.037	0.036
S5	0.050	0.024	0.10	0.13	0.045	0.029	0.078	0.092
S6	0.042	0.019	0.085	0.098	0.034	0.023	0.059	0.065
S7	0.037	0.017	0.081	0.11	0.032	0.020	0.055	0.064
S8	0.037	0.019	0.087	0.11	0.038	0.026	0.072	0.079
S9	0.043	0.020	0.098	0.12	0.040	0.025	0.071	0.081
S10	0.027	0.012	0.064	0.078	0.026	0.013	0.047	0.051
E1	0.004	0.004	0.008	0.006	0.007	0.007	0.009	0.009
E2	0.015	0.007	0.041	0.051	0.018	0.013	0.033	0.038
E3	0.007	0.004	0.009	0.009	0.009	0.006	0.008	0.008
E4	0.005	0.003	0.007	0.007	0.006	0.005	0.006	0.005
E5	0.043	0.010	0.071	0.090	0.029	0.021	0.045	0.058
E6	0.017	0.011	0.059	0.082	0.035	0.026	0.057	0.075
E7	0.022	0.010	0.053	0.074	0.024	0.017	0.041	0.053
E8	0.025	0.011	0.073	0.092	0.030	0.020	0.055	0.063
E9	0.030	0.012	0.078	0.11	0.033	0.023	0.054	0.070
E10	0.016	0.009	0.043	0.061	0.021	0.016	0.037	0.039

Site	B[b]Fl	B[e]P	B[a]P	Per	Ind	B[ghi]P	T-PAH	IL(%)
D1	6.8	3.1	4.0	1.2	2.8	2.5	35.8	
D2	6.9	3.1	4.0	1.1	2.6	2.3	36.6	
D3	0.80	0.46	0.56	0.31	0.40	0.38	5.57	
K1	0.92	0.51	0.59	0.30	0.42	0.45	6.60	
S1	0.36	0.12	0.19	0.049	0.15	0.14	1.80	5.6
S2	0.22	0.078	0.12	0.034	0.093	0.081	1.09	4.9
S3	0.18	0.062	0.081	0.025	0.071	0.066	0.884	4.9
S4	0.10	0.037	0.050	0.015	0.037	0.038	0.522	5.8
S5	0.22	0.076	0.11	0.033	0.089	0.083	1.16	4.6
S6	0.15	0.052	0.079	0.022	0.063	0.056	0.842	5.5
S7	0.16	0.059	0.080	0.023	0.069	0.062	0.870	4.9
S8	0.17	0.055	0.091	0.026	0.067	0.039	0.912	5.1
S9	0.21	0.072	0.11	0.031	0.086	0.077	1.08	5.7
S10	0.15	0.052	0.073	0.022	0.066	0.060	0.736	5.8
E1	0.014	0.008	0.011	0.008	0.009	0.014	0.117	4.9
E2	0.064	0.024	0.042	0.013	0.037	0.036	0.431	3.5
E3	0.013	0.007	0.009	0.007	0.011	0.010	0.115	3.5
E4	0.008	0.005	0.006	0.005	0.008	0.008	0.083	11.9
E5	0.099	0.038	0.058	0.016	0.045	0.041	0.664	10.0
E6	0.13	0.051	0.075	0.020	0.063	0.057	0.760	22.0
E7	0.10	0.039	0.051	0.017	0.045	0.042	0.591	21.8
E8	0.12	0.048	0.069	0.018	0.058	0.052	0.735	10.0
E9	0.13	0.047	0.069	0.019	0.062	0.059	0.786	5.1
E10	0.054	0.030	0.046	0.013	0.040	0.036	0.462	4.1

T-PAH : total amounts of 14 PAH.
IL : ignition loss.

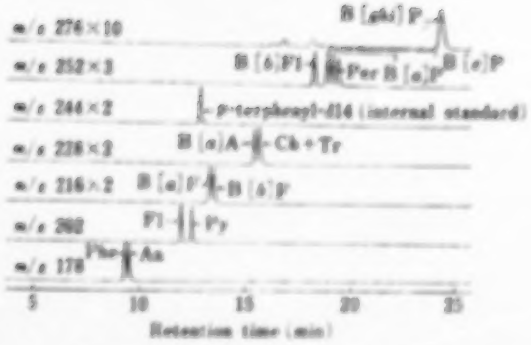

Fig. 2. SIM Chromatogram of PAH

Table IV. Values of Factor Loading and Eigenvalues of Z1 and Z2

PAH	Z1	Z2
Ph	0.7510	−0.5429
An	0.9618	−0.0997
Fl	−0.4866	−0.7352
Py	−0.5586	0.5040
B[a]F	0.8514	−0.4214
B[b]F	0.9796	−0.7217
B[a]A	0.9165	−0.4160
Ch+Tr	0.6649	−0.1865
B[k]Fl	−0.9831	0.3776
B[e]P	0.7093	0.5547
B[a]P	0.1749	0.6723
Per	0.9669	0.0562
Ind	−0.5250	0.5537
B[ghi]P	0.6369	0.6929
Eigenvalue	7.324	3.713
RC	0.523	0.239
CUM. RC	0.523	0.753

RC : ratio of contribution
CUM. RC : cumulative ratio of contribution

Table V. Component Scores of Z1 and Z2

site	Z1	Z2
H1	0.721	−0.639
H2	0.726	1.028
H3	7.239	2.934
K1	4.753	−0.661
S1	−1.653	1.433
S2	−1.433	2.266
S3	−1.052	−0.141
S4	−1.323	−0.610
S5	−0.569	−0.851
S6	−0.728	−2.695
S7	−1.369	−1.086
S8	−1.751	−3.348
S9	−1.284	−0.167
S10	−1.912	1.928

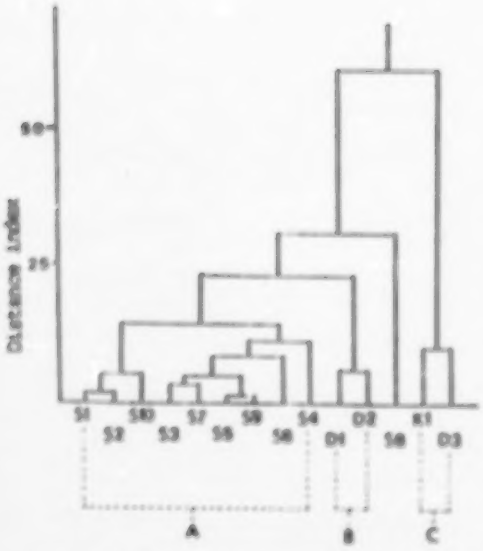

Fig. 3. Dendrogram Comparing Sample Sites

Bibliography

UNEDITED TRANSLATION
For information only
TRADUCTION NON REVISEE
Information seulement

引 用 文 献

1) F.G. Prahl, E.C. Cercellus, T. Carpenter, *Environ. Sci. Technol.*, 18, 687 (1984); R. Shinohara, M. Koga, A. Kido, S. Eto, T. Tabata, T. Hori, T. Akiyama, *Environment International*, 4, 163 (1980); 篠原亮一, 来田静太郎, 重森伸雄, 秋山 高, 大気汚染学会誌, 14, 143 (1979).
2) 篠原亮太, 古賀 実, 堀 勉二, 秋山 高, 衛生化学, 24, 304 (1978).
3) L. Martel, J.M. Gagnon, R. Masse, A. Leclerc, *Water Res.*, 21, 699 (1987).
4) 北九州市環境衛生研究所, 昭和53年度化学物質環境追跡調査結果報告(環境庁委託研究), 1979, p. 2.
5) 河原純明, 横山ひろみ, 桑山 登, 白井文雄, 衛生化学, 34, 248 (1988).
6) T.D. Searl, F.J. Cassidy, W.H. King, R.A. Brown, *Anal. Chem.*, 42, 954 (1970).
7) J.C. Means, *Environ. Sci. Technol.*, 14, 1524 (1980).
8) 伊 順子, 石渡良志, 地球化学, 17, 53 (1983).
9) 田中 豊, 垂水共之, 脇本和昌(編), "パソコン統計解析ハンドブックII 多変量解析編," 共立出版, 東京, 1984, p.236.
10) 奥野忠一, 芳賀敏郎, 久米 均, 古澤 正, "多変量解析法," 改訂版, 日科技連, 東京, 1981, p.159.

1) Tetsuichi TABATA, Shintaro SUETA, Nobuyasu SHIGEMORI, Takashi AKIYAMA : Taiki Osen Gakkaishi (Journal of the Society for Atmospheric Studies), 14, 143 (1979)

2) Ryota SHINOHARA, Minoru KOGA, Teiji HORI, Takashi AKIYAMA : this journal, 24, 304 (1978)

4) Kitakyushu-shi Kankyo Eisei Kenkyusho (Kitakyushu Municipal Research Center for Environmental Health), Showa 53 nendo Kagaku Busshitsu Kankyo Tsuiseki Chosa Kekka Hokoku (1978 Report on the Results obtained through Investigations regarding Chemical Substances and the Environment) (Study commissioned by the Environment Agency), 1979, p. 2

5) Kuniaki KAWADA, Hiromi YOKOYAMA, Noboru MORIYAMA, Fumio SHIRAI : this journal, 34, 954 (1970)

8) Junko IN, Ryoji ISHIWATARI, Chikyu Kagaku (Geochemistry), 17, 53 (1983)

9) Yutaka TANAKA, Tomoyuki TARUMI, Kazumasa WAKIMOTO (Ed.) : "Pasokon Tokei Kaiseki Handobukku II Tahenryo Kaiseki-hen" (Personal Computer Statistics

Analysis Handbook II, Multivaried Analysis part", Kyoritsu Publ., Tokyo, 1984, p. 226

10) Chuichi OKUNO, Toshiro HAGA, Hitoshi KUME, Tadashi FURUZAWA, "Tahenryo Kaiseki-ho" ("Multivaried Analysis Method"), Revised Edition, Nikka Giren Publ., Tokyo, 1981, p. 159